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Deposition of ^{125}Sb, ^{106}Ru, ^{144}Ce, ^{134}Cs and ^{137}Cs in Finland after the Chernobyl accident

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In this study the deposition characteristics of ^{125}Sb, ^{106}Ru, ^{144}Ce, ^{134}Cs and ^{137}Cs in Finland after the Chernobyl accident was investigated based on gamma spectrometric analysis of 97 lichen, peat and surface soil samples. The aim of this study was to determine the fallout pattern of ^{125}Sb, ^{106}Ru, ^{134}Cs, and ^{144}Ce in Finland, to verify the fallout pattern of ^{137}Cs reported in earlier works, and to obtain an estimate of the total amounts of these nuclides deposited in Finland. The highest deposition values of ^{144}Ce were found in a zone extending from southwestern Finland to Kuhmo area close to the Russian border. The deposition pattern of ^{144}Ce resembled the deposition patterns of refractory nuclides, such as ^{95}Zr and transuranium elements. The regional deposition of cesium isotopes 134 and 137 differed from that of ^{144}Ce owing to the different volatility properties of these nuclides. Our results confirm the earlier observations of high deposition values of ^{137}Cs in southwestern Finland, Varkaus–Kuopio region, Kuhmo region and Kotka–Kouvola region. A comparison of previous results to our results suggests an overestimated deposition in the Oulu region in this study due to the lack of samples in the region. The observed average ^{134}Cs to ^{137}Cs activity ratio, 0.527 ± 0.010 decay-corrected to 1 May 1986, is in agreement with earlier studies and corresponds to a burnup of 13 GWd tU^{-1}. Although ruthenium is a refractory element, the behaviour of ^{103}Ru and ^{106}Ru has been shown to resemble the behaviour of volatile elements. This has been explained by the formation of volatile ruthenium oxides. The deposition pattern of ^{125}Sb resembled those of cesium isotopes. This suggests that antimony behaved like volatile nuclides in the destroyed reactor. It was calculated that depending on the nuclide, 0.017%–1.5% of the reactor core inventory and 0.6%–13% of the atmospheric emissions were deposited in Finland. These percentages were proportional to the volatility of the nuclides.

Introduction

The worst ever peace-time nuclear accident happened in the former Soviet Union on 26 April 1986. An accident destroyed the Chernobyl number four reactor, leading to a rapid loss of over 30 human lives and to a significant radioactive contamination of the environment.

According to the Soviet estimates, all the radioactive noble gases of the reactor core inventory were liberated during the accident. 10%–20% of the volatile nuclides, e.g. ^{131}I and ^{137}Cs, were distributed into the environment. A fraction (2%–6%) of refractory nuclides, such as ^{95}Zr and transuranium elements, was released as well (USSR State Committee on the Utilization of Atomic Energy 1986). Dose calculations relating to the accident have been published by Aarkrog (1988) and Bouville (1995).

The energy released in the hydrogen explosion and the heat produced by burning graphite and decaying fission products caused the radioactive debris to reach considerable altitudes with correspondingly high wind speeds. This, in turn, caused the debris to spread quickly in the atmosphere. At first the emissions were transported northwestwards to Poland, the Baltic states, Sweden, Norway and Finland. During 27 April, emissions spread to eastern-central Europe, southern Germany, Italy and Yugoslavia. During the next week the plume was transported southwards from Chernobyl to Romania, Bulgaria, the Balkans, the Black Sea and Turkey. The emissions of 5 May arrived again over central Europe, Scandinavia and Finland during 6–9 May (Persson *et al*. 1987). Most of the Chernobyl-originated activity remained in the troposphere. Jaworowski and Kownacka (1988) reported that the stratospheric activities were only 1%–6% of those found in the surface air. Eventually the plume was distributed practically all over the northern hemisphere, its advance being monitored both by measurements and by air mass trajectory calculations.

According to the calculations of Valkama *et al*. (1995), the air parcel trajectories originating from Chernobyl at the time of the accident arrived at Finland from the south-west at the release heights of 1500 and 2500 metres. Their arrival time in south-west Finland was 12:00 UTC on 27 April 1986 for the release height of 2000 m and the arrival height of 1500 m, and 15:00 UTC on 27 April for the release height of 1500 m and the arrival height of 1000 m. Aircraft measurements made over the southern Finland daily starting on 28 April are in agreement with these arrival heights (Sinkko *et al*. 1987).

After the first few days no significant amount of radioactivity was deposited in Finland, even though somewhat higher values were observed on 11 and 13 May in southern Finland. In Nurmijärvi, southern Finland, the concentration of ^{137}Cs in the ground-level air decreased by four orders of magnitude between 28 April and 16 May, 1986, starting from the value of 10^4 mBq m^{-3} (Finnish Centre for Radiation and Nuclear Safety 1986a, 1986b).

In the present study a number of ^{125}Sb, ^{106}Ru, ^{144}Ce, ^{134}Cs and ^{137}Cs deposition values following the Chernobyl accident were collected. They are based on a gamma-spectrometric analysis of peat, lichen and soil samples. The regional distribution of these deposition values was studied. The aims of this study was (1) to find out the fallout patterns of ^{125}Sb, ^{106}Ru, ^{134}Cs and ^{144}Ce after the Chernobyl accident, (2) to verify the fallout pattern of ^{137}Cs reported in the earlier works and (3) to obtain an estimate of the total amount of these nuclides deposited in Finland.

Experimental and computational methods

In May 1986, the National Public Health Institute collected 62 peat samples from peatlands in production. The sampling areas had been untouched since the peat harvesting season of 1985. A detailed description of the sampling procedure and the results of gamma spectrometric analyses have been published by Jantunen *et al*. (1987) and Jantunen *et al*. (1991). At the Department of Radiochemistry, University of Helsinki, the peat samples were analysed again using the gamma spectrometry followed by radiochemical separations to analyse their contents of transuranium nuclides (Reponen *et al*. 1993, Paatero *et al*. 1994a, Salminen *et al*. 2005).

Most of the lichen samples analyzed in this study were collected in 1986 by the staff of the Department of Radiochemistry, University of Helsinki. The sampling of lichen was usually performed by collecting all the lichen from a definite area. These lichen-carpet profile samples were air-dried in the laboratory and usually divided into four fractions: the upper and

lower parts of the lichen, litter and underlying soil. One sample was collected in 1987. Some additional lichen samples were obtained from the Finnish Centre for Radiation and Nuclear Safety.

The peat and lichen samples were analysed using the HPGe gamma spectrometry. The lichen samples were analysed in 1986 and the peat samples were analysed in 1989. Thus the gamma emitters with a half life less than a few months had already decayed from the peat samples. The spectra were handled with the MicroSAMPO peak fit program (Aarnio *et al*. 1988).

Finnish Centre for Radiation and Nuclear Safety collected about 10 surface soil samples around Finland in 1986 and analysed them with the semiconductor gamma-spectrometry. The activity concentrations of the radionuclides were calculated using the computer program GAMMA-83 (Saxén *et al*. 1987).

The cases of missing results owing to concentrations below minimum detectable activity were treated in the subsequent analysis as follows: The missing values were replaced with a value that was one fourth of the lowest observed value within the corresponding sample type (peat, soil, lichen). It was assumed that the lowest deposition was twice the minimum detectable activity. In atmospheric sciences it is a common practice to replace the below-detection-limit values with one half of the detection-limit value. Altogether this yielded 97 deposition values, mainly in southern and central Finland.

The deposition values were extrapolated over Finland by taking into account the amount of precipitation causing wet deposition. The sums of daily precipitation amounts between 27 April and 17 May 1986 were calculated at 591 meteorological and hydrological monitoring stations in Finland. This time period was chosen to cover the occurrence of most of the Chernobyl fallout based on daily total beta activity deposition (Paatero *et al*. 1994b). The deposition values were divided by the amount of precipitation at the sampling site in order to get the concentration values in precipitation. The precipitation estimates were interpolated from all the 591 precipitation amounts using inverse distance weighing (IDW):

$$\hat{C}_j = \frac{\sum_{i=1}^{n} w_i C_i}{\sum_{i=1}^{n} w_i}. \quad (1)$$

Here $\hat{C}_j$ denotes the estimated quantity (precipitation amount in this case) at the location of interest j, C_i the observed quantity at the location i, n is the number of observations and w_i is the weight factor related to the distance $d_{i,j}$ between the locations i and j and is given by

$$w_i = \frac{1}{d_{i,j}^3}. \quad (2)$$

The distances $d_{i,j}$ were calculated using the following equation:

$$d_{i,j} = R \arccos[\sin\phi_j \times \sin\phi_i + \cos\phi_j \times \cos\phi_i \times \cos(\lambda_j - \lambda_i)]. \quad (3)$$

Here R is the radius of the Earth (6371 km), and ϕ and λ are the latitude and longitude of the location, respectively, when assuming a spherical Earth (Bonham-Carter 1996). The use of weights that are inversely proportional to the third power of the distance strongly enhances the significance of the closest observations.

Next a concentration value grid between the latitudes 60°N and 70°N and the longitudes 20°E and 31°E was calculated using the IDW method. The resolution of the calculated grid was 0.5 degrees in the north–south direction and 1.0 degree in the east–west direction. A similar grid was calculated for the precipitation amounts. A deposition estimate was calculated at each grid node by multiplying the concentration value by the precipitation amount (Paatero *et al*. 2002).

Results and discussion

Figure 1 shows the locations of the regions. The observed deposition values of ^{125}Sb, ^{106}Ru, ^{144}Ce, ^{134}Cs and ^{137}Cs based on peat and lichen samples are reported in Tables 1–4. The deposition values obtained with the soil samples have been reported in the work of Saxén *et al*. (1987). Statistical parameters of the interpolated deposition values are presented in Table 5.

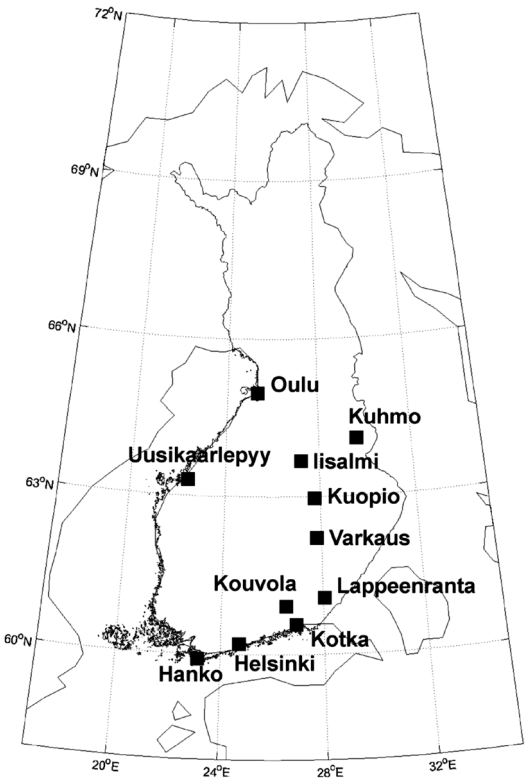

Fig. 1. Studied locations.

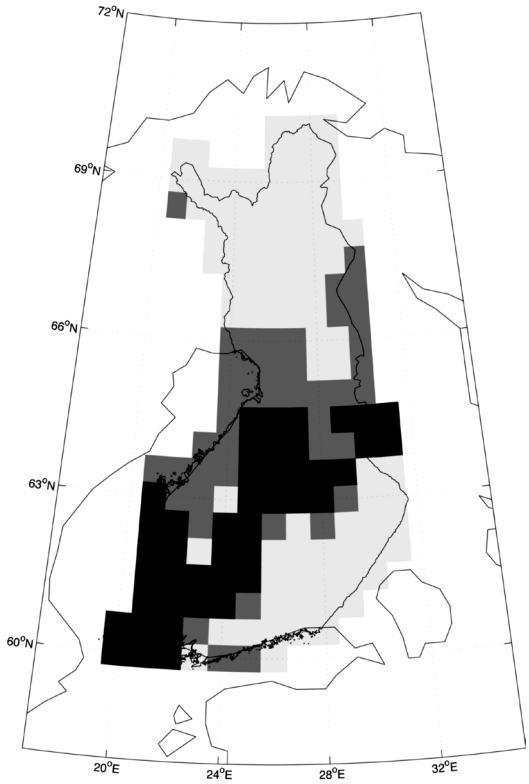

Fig. 2. Regional distribution of the ^{144}Ce deposition (Bq m^{-2}) in Finland decay-corrected to 1 May 1986. The three classes of deposition represent the 0–50 (light grey, < 647 Bq m^{-2}), 50–75 (dark grey, 647–2159 Bq m^{-2}) and 75–100 (black, > 2159 Bq m^{-2}) percentiles of the interpolated deposition values.

The highest deposition values of ^{144}Ce were found in a zone extending from southwestern Finland to the Kuhmo area close to the Russian border (Fig. 2). The deposition pattern of ^{144}Ce resembled the deposition patterns of refractory nuclides such as ^{95}Zr and transuranium elements (Arvela *et al*. 1990, Paatero *et al*. 2002, Salminen *et al*. 2005). The highest ^{144}Ce deposition values were found along the calculated trajectory of the air mass which was located in the Chernobyl region during the initial explosion of the reactor (Valkama *et al*. 1995). The deposition was high

Table 1. The observed deposition values (Bq m^{-2} ± 1 σ propagated counting error decay-corrected to 1 May 1986) of ^{134}Cs and ^{137}Cs based on peat samples.

No.	Community	Lat. (°N)	Long. (°E)	Cs-134		Cs-137	
				(Bq m^{-2})	± 1 σ	(Bq m^{-2})	± 1 σ
4	Karvia	62.0	22.7	8300	200	15200	500
10	Kankaanpää	61.8	22.8	11500	150	19400	600
11	Kankaanpää	61.7	22.8	35800	500	59600	1900
14	Tampere	61.3	23.9	111900	1500	188000	7000
15	Punkalaidun	61.0	23.2	11900	140	20100	700
16	Köyliö	61.0	22.5	15900	200	26600	900
17	Hattula	60.7	24.2	13400	300	25200	800
18	Loimaan mlk	60.7	23.0	25300	300	43100	1400

continued

Table 1. Continued.

No.	Community	Lat. (°N)	Long. (°E)	Cs-134		Cs-137	
				(Bq m^{-2})	± 1 σ	(Bq m^{-2})	± 1 σ
19	Janakkala	60.8	24.7	12100	200	20300	700
20	Riihimäki	60.6	24.7	20100	300	33100	1100
21	Hankasalmi	62.2	26.3	7850	140	13300	500
22	Leivonmäki	61.7	25.9	4050	90	6900	200
25	Viitasaari	63.1	25.8	5260	50	8900	300
26	Pihtipudas	63.3	25.5	8040	140	13900	600
27	Heinolan mlk	61.0	25.9	2690	60	4670	160
36	Kyyjärvi	62.8	24.6	12000	160	20700	700
37	Juva	61.8	27.2	2530	50	4370	140
40	Joutseno	60.9	28.4	280	10	490	20
41	Rautjärvi	61.2	29.0	390	14	700	30
42	Punkaharju	61.5	29.0	1080	30	1940	70
44	Taipalsaari	61.2	28.0	840	20	1520	50
45	Valkeala	60.9	27.1	19400	200	32400	1000
46	Haukivuori	61.9	27.1	8600	120	14400	500
47	Mikkelin mlk	61.7	27.2	410	11	720	30
48	Kontiolahti	62.7	29.3	327	7	610	20
49	Tohmajärvi	62.3	29.8	270	9	480	20
50	Ilomantsi	62.8	30.6	112	6	219	9
51	Tuupovaara	62.5	30.0	213	7	670	20
52	Pyhäselkä	62.4	29.7	411	12	720	30
53	Rääkkylä	62.2	29.5	429	12	760	30
54	Rautavaara	63.3	28.2	1390	20	2570	80
55	Suonenjoki	62.6	26.6	2640	60	4470	150
55	Suonenjoki	62.6	26.6	1860	40	3140	110
56	Suonenjoki	62.7	27.2	10200	110	17000	600
57	Pielavesi	63.1	26.8	7160	110	12300	400
58	Karttula	62.8	26.8	5860	130	9900	300
60	Kiuruvesi	63.3	26.3	2540	90	8800	300
61	Kiuruvesi	63.6	26.3	2110	30	3630	120
62	Kiuruvesi	63.7	26.4	2540	40	4600	200
63	Kiuruvesi	63.5	26.3	4240	70	7900	260
64	Sonkajärvi	63.8	27.2	4120	80	8600	300
65	Ilomantsi	62.9	30.6	105	4	434	15
95	Vimpeli	63.1	24.0	17700	190	28700	900
96	Halsua	63.2	24.5	10700	300	18000	600
99	Jalasjärvi	62.3	22.9	13300	200	22200	700
100	Peräseinäjoki	62.4	23.2	3500	70	6100	200
110	Köyliö	61.0	22.4	8330	110	14200	500
111	Loimaa	60.7	23.1	7080	100	12300	400
117	Hartola	61.3	25.9	12400	200	20800	700
135	Kerimäki	61.9	28.8	6100	100	10400	400
136	Mikkelin mlk	61.8	27.2	3520	80	6000	200
140	Lieksa	63.3	29.8	197	5	1010	30
143	Valtimo	63.7	28.4	2700	60	5100	200
144	Rautavaara	63.5	27.6	2600	60	4820	160
146	Suonenjoki	62.4	27.1	16500	300	27200	900
148	Pielavesi	63.0	26.8	10300	160	17500	600
149	Keitele	63.0	26.1	7400	70	12200	500
152	Keitele	63.2	26.1	2990	50	5200	200
168	Kauhava	63.0	23.5	14400	200	26200	900
201	Kitee	62.2	29.6	343	7	720	20
202	Lieksa	63.2	30.0	157	4	540	20

Table 2. The observed deposition values (Bq m^{-2} ± 1 σ propagated counting error decay-corrected to 1 May 1986) of ^{125}Sb, ^{106}Ru, and ^{144}Ce based on peat samples (n.d. = not detected).

No.	Community	Ru-106		Sb-125		Ce-144	
		(Bq m^{-2})	± 1 σ	(Bq m^{-2})	± 1 σ	(Bq m^{-2})	± 1 σ
4	Karvia	n.d.		n.d.		n.d.	
10	Kankaanpää	19500	1200	620	20	2200	200
11	Kankaanpää	40000	3000	1680	80	n.d.	
14	Tampere	n.d.		5200	300	33000	7000
15	Punkalaidun	14900	400	625	14	5700	200
16	Köyliö	28000	1400	900	50	7400	600
17	Hattula	n.d.		n.d.		n.d.	
18	Loimaan mlk	15000	3000	1180	40	n.d.	
19	Janakkala	n.d.		630	60	n.d.	
20	Riihimäki	15000	2000	850	30	n.d.	
21	Hankasalmi	n.d.		n.d.		n.d.	
22	Leivonmäki	n.d.		140	40	n.d.	
25	Viitasaari	7300	600	229	8	3500	200
26	Pihtipudas	46000	3000	330	20	5400	500
27	Heinolan mlk	n.d.		126	11	n.d.	
36	Kyyjärvi	n.d.		360	50	3000	600
37	Juva	n.d.		130	20	n.d.	
40	Joutseno	n.d.		n.d.		n.d.	
41	Rautjärvi	2100	500	n.d.		n.d.	
42	Punkaharju	4700	1300	n.d.		n.d.	
44	Taipalsaari	n.d.		n.d.		n.d.	
45	Valkeala	15000	3000	860	40	n.d.	
46	Haukivuori	8200	1500	360	20	n.d.	
47	Mikkelin mlk	n.d.		n.d.		n.d.	
48	Kontiolahti	2100	300	30	2	340	90
49	Tohmajärvi	n.d.		n.d.		n.d.	
50	Ilomantsi	n.d.		n.d.		n.d.	
51	Tuupovaara	n.d.		n.d.		n.d.	
52	Pyhäselkä	n.d.		n.d.		n.d.	
53	Rääkkylä	n.d.		n.d.		n.d.	
54	Rautavaara	3700	700	n.d.		2400	200
55	Suonenjoki	n.d.		n.d.		n.d.	
55	Suonenjoki	n.d.		n.d.		n.d.	
56	Suonenjoki	12600	1100	390	15	1800	300
57	Pielavesi	n.d.		n.d.		5300	600
58	Karttula	n.d.		270	50	2900	600
60	Kiuruvesi	73000	3000	230	20	7100	400
61	Kiuruvesi	16000	500	85	8	2500	200
62	Kiuruvesi	5200	900	107	5	3500	200
63	Kiuruvesi	17000	2000	240	20	11400	600
64	Sonkajärvi	12000	2000	n.d.		8500	600
65	Ilomantsi	n.d.		n.d.		n.d.	
95	Vimpeli	13900	900	680	30	n.d.	
96	Halsua	n.d.		n.d.		n.d.	
99	Jalasjärvi	n.d.		590	30	6300	600
100	Peräseinäjoki	3400	800	144	13	800	200
110	Köyliö	11600	800	460	20	3500	400
111	Loimaa	15100	900	390	20	n.d.	
117	Hartola	n.d.		460	40	n.d.	
135	Kerimäki	3100	600	270	9	n.d.	
136	Mikkelin mlk	7400	1800	230	20	n.d.	
140	Lieksa	n.d.		n.d.		n.d.	

continued

Table 2. Continued.

No.	Community	Ru-106		Sb-125		Ce-144	
		(Bq m^{-2})	± 1 σ	(Bq m^{-2})	± 1 σ	(Bq m^{-2})	± 1 σ
143	Valtimo	n.d.		n.d.		11200	1100
144	Rautavaara	27000	3000	n.d.		8400	600
146	Suonenjoki	n.d.		480	90	n.d.	
148	Pielavesi	n.d.		390	40	4400	800
149	Keitele	11900	1000	315	11	8500	300
152	Keitele	4700	1000	130	12	4400	300
168	Kauhava	n.d.		490	30	1600	400
201	Kitee	n.d.		28	6	n.d.	
202	Lieksa	1900	300	19	4	n.d.	

Table 3. The observed deposition values (Bq m^{-2} ± 1 σ propagated counting error decay-corrected to 1 May 1986) of ^{134}Cs and ^{137}Cs based on lichen samples.

Sample no.	Community	Lat. (°N)	Long. (°E)	Cs-134		Cs-137	
				(Bq m^{-2})	± 1 σ	(Bq m^{-2})	± 1 σ
300-86	Vaskijärvi	60.6	24.3	3550	50	6800	200
302-86	Jokioinen	60.8	23.5	4030	50	7200	200
303-86	Kalanti, Jaakkola	60.8	21.5	46500	300	75500	1500
304-86	Uusikaupunki	60.8	21.4	37100	200	61300	1300
304-86b	Uusikaupunki	60.8	21.4	33500	400	58000	2000
306-86	Kalanti, kirkko	60.8	21.6	46600	400	86000	2000
308-86	Kalanti, kirkko	60.8	21.6	38900	200	62900	1300
310-86	Hiidenvesi	60.4	24.3	1380	20	2720	60
311-86	Yläne	60.9	22.3	29400	300	50900	1000
314-86	Wessö	60.3	25.8	7730	70	15400	200
316-86	Pietarsaari	63.7	22.7	21300	200	36300	800
317-86	Wessö	60.3	25.8	9990	130	20900	500
318-86	Wessö	60.3	25.8	4420	50	10000	200
321-86	Wessö	60.3	25.8	4910	50	10700	200
322-86	Wessö	60.3	25.8	6090	90	12500	200
323-86	Pärnäsaari	61.2	25.6	41600	200	64700	1200
325-86	Tenhola	60.0	23.2	825	10	2160	30
326-86	Båtviken	59.9	23.3	440	6	1900	30
327-86	Ristiina	61.5	27.3	4300	50	8390	140
328-86	Huittinen	61.2	22.8	27000	200	45800	900
330-86	Wessö	60.3	25.8	4440	50	8300	200
337-86	Kausala	60.9	26.3	43600	400	70000	1400
338-86	Pertunmaa	61.4	26.5	8840	90	16400	500
421-87	Sotkamo	64.0	28.8	2030	30	6840	140
10-87	Kaamanen	69.1	27.1	377	9	1320	50

in areas with simultaneous occurrence of precipitation (Paatero *et al*. 2002). On the other hand, a high deposition occurred at the southwestern coast of Finland despite the lack of precipitation in this area. This can be attributed to the dry deposition of hot particles (Saari *et al*. 1989). The aerodynamic diameters of these particles were of such magnitude that their gravitational settling had to be taken into account when assessing their behaviour in the atmosphere (Pöllänen *et al*. 1997). The deposition might have been overestimated in the region of Oulu because there were no samples from this area. A relatively low deposition occurred in the Hanko region. This

Table 4. The observed deposition values (Bq m^{-2} ± 1 σ propagated counting error decay-corrected to 1 May 1986) of ^{125}Sb, ^{106}Ru, and ^{144}Ce based on lichen samples.

Sample no.	Community	Ru-106		Sb-125		Ce-144	
		(Bq m^{-2})	± 1 σ	(Bq m^{-2})	± 1 σ	(Bq m^{-2})	± 1 σ
300-86	Vaskijärvi	1650	60	133	9	240	20
302-86	Jokioinen	1820	50	159	8	760	20
303-86	Kalanti, Jaakkola	16900	300	1930	40	9000	200
304-86	Uusikaupunki	14400	200	1650	30	2920	90
304-86b	Uusikaupunki	11400	400	1300	70	3050	130
306-86	Kalanti, kirkko	29100	500	2590	60	3480	100
308-86	Kalanti, kirkko	14700	300	1650	40	2950	80
310-86	Hiidenvesi	540	20	60	5	238	11
311-86	Yläne	13500	200	1320	30	5060	110
314-86	Wessö	5270	110	298	11	660	30
316-86	Pietarsaari	1580	60	510	20	450	30
317-86	Wessö	6900	200	440	20	720	50
318-86	Wessö	2980	80	118	9	520	30
321-86	Wessö	4070	100	590	50	204	9
322-86	Wessö	3270	140	230	30	150	20
323-86	Pärnäsaari	2970	80	840	20	870	30
325-86	Tenhola	470	20	n.d.		91	5
326-86	Båtviken	770	60	17	2	235	9
327-86	Ristiina	950	30	105	6	160	9
328-86	Huittinen	9300	200	970	30	3430	90
330-86	Wessö	2790	70	117	7	n.d.	
337-86	Kausala	3660	100	830	30	370	20
338-86	Pertunmaa	1490	40	94	3	188	8
421-87	Sotkamo	1320	40	n.d.		1090	30
10-87	Kaamanen	n.d.		n.d.		n.d.	

Table 5. Statistical parameters of the interpolated deposition values (Bq m^{-2} decay-corrected to 1 May 1986).

Parameter	Cs-134	Cs-137	Ru-106	Sb-125	Ce-144
Minimum	38	97	35	1	19
10% value	522	1104	323	13	148
25% value	1103	2148	882	36	270
Median	2450	4806	2022	96	647
75% value	8312	15032	5346	330	2159
90% value	20155	35940	13213	927	4209
Maximum	108980	183140	28148	5216	30952
Arithm. mean	7319	12864	4202	319	1615
Geom. mean	2966	5677	2002	105	706

can be explained by the surrounding cold sea surface which stratifies the lower troposphere, thereby reducing the vertical transport of radionuclides. On the other hand, this implies that the obtained deposition value for the south-western archipelago was probably overestimated.

The regional deposition of cesium isotopes 137 and 134 (Figs. 3 and 4) differed from that of ^{144}Ce because cesium is a volatile element with a boiling point of 678 °C. Compared with the refractory nuclides, a higher proportion of cesium was emitted from the burning reactor during several days after the initial explosion. The deposition was highest in southwestern Finland. Again, the deposition was relatively low in the Hanko–Helsinki region. Our results confirm the earlier *in situ* observations of high deposition values in the Varkaus–Kuopio region, Kuhmo

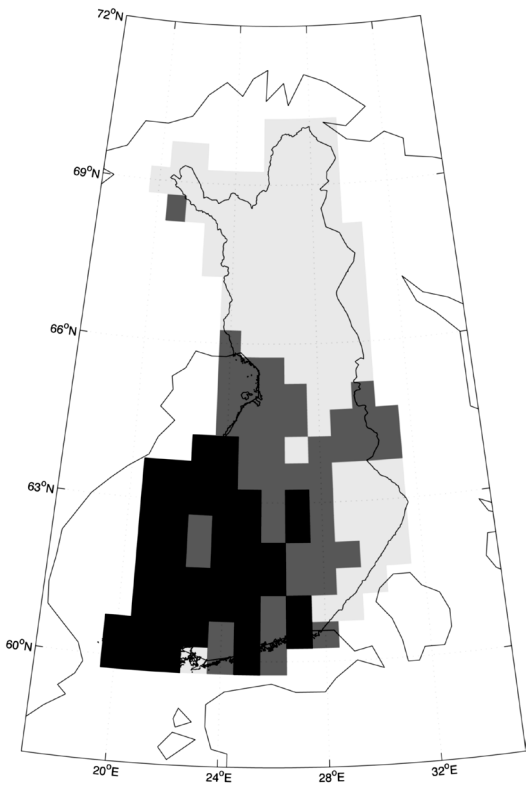

Fig. 3. Regional distribution of the ^{137}Cs deposition (Bq m^{-2}) in Finland decay-corrected to 1 May 1986. The three classes of deposition represent the 0–50 (light grey, < 4806 Bq m^{-2}), 50–75 (dark grey, 4806–15032 Bq m^{-2}) and 75–100 (black, > 15032 Bq m^{-2}) percentiles of the interpolated deposition values.

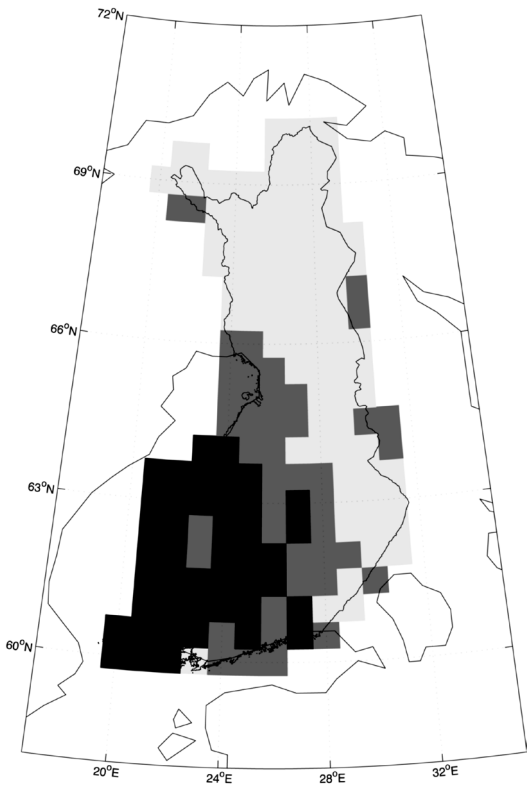

Fig. 4. Regional distribution of the ^{134}Cs deposition (Bq m^{-2}) in Finland decay-corrected to 1 May 1986. The three classes of deposition represent the 0–50, 50–75, and 75–100 percentiles of the interpolated deposition values. The three classes of deposition represent the 0–50 (light grey, < 2450 Bq m^{-2}), 50–75 (dark grey, 2450–8312 Bq m^{-2}) and 75–100 (black, > 8312 Bq m^{-2}) percentiles of the interpolated deposition values.

region and Kotka–Kouvola region (Arvela *et al*. 1990, Kettunen 2006). A comparison of previous results with our results suggests that an overestimated deposition in the Oulu region was obtained in this study. The deposition values obtained in this study are of the same order of magnitude as those found in Sweden, ranging from a few kBq m^{-2} to > 85 kBq m^{-2} (Persson *et al*. 1987). In Estonia, the ^{137}Cs deposition varied between 0.11 and 50 kBq m^{-2} (Lust *et al*. 2006). The highest deposition values were found in northeastern Estonia. This is in agreement with our results, showing that the deposition was high in the Kotka–Kouvola region. This area is directly north of the northeastern Estonia on the northern shore of the Gulf of Finland. The observed average ^{134}Cs to ^{137}Cs activity ratio, 0.527 ± 0.010 decay-corrected to 1 May 1986, is in agreement with the earlier studies. Arvela *et al*. (1989) reported a value of 0.59 for this ratio, while Antonov *et al*. (1988) gave a value of 0.486 and Toivonen *et al*. (1988) gave a value of 0.55 ± 0.09. Early Soviet emission estimates gave a slightly higher value of 0.655 (International Atomic Energy Agency 1986). Based on the core inventory calculations of Kirchner and Noack (1988), the ratio observed in this study corresponds to a burnup of 13 GWd tU^{-1}, also in agreement with the earlier studies (USSR State Committee on the Utilization of Atomic Energy 1986, Persson *et al*. 1987, Paatero *et al*. 1998).

Even though ruthenium is a refractory element, the behaviour of ^{103}Ru and ^{106}Ru has been shown to resemble the behaviour of volatile ele-

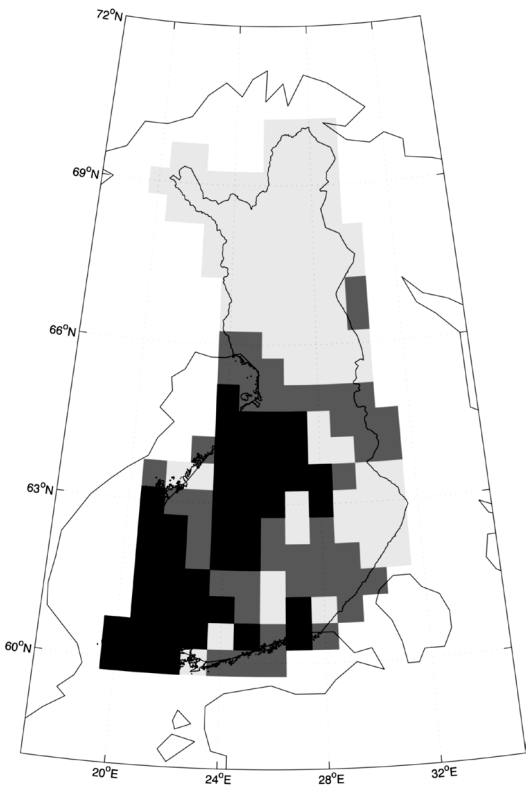

Fig. 5. Regional distribution of the ^{106}Ru deposition (Bq m^{-2}) in Finland decay-corrected to 1 May 1986. The three classes of deposition represent the 0–50, 50–75, and 75–100 percentiles of the interpolated deposition values. The three classes of deposition represent the 0–50 (light grey, < 2022 Bq m^{-2}), 50–75 (dark grey, 2022–5346 Bq m^{-2}) and 75–100 (black, > 5346 Bq m^{-2}) percentiles of the interpolated deposition values.

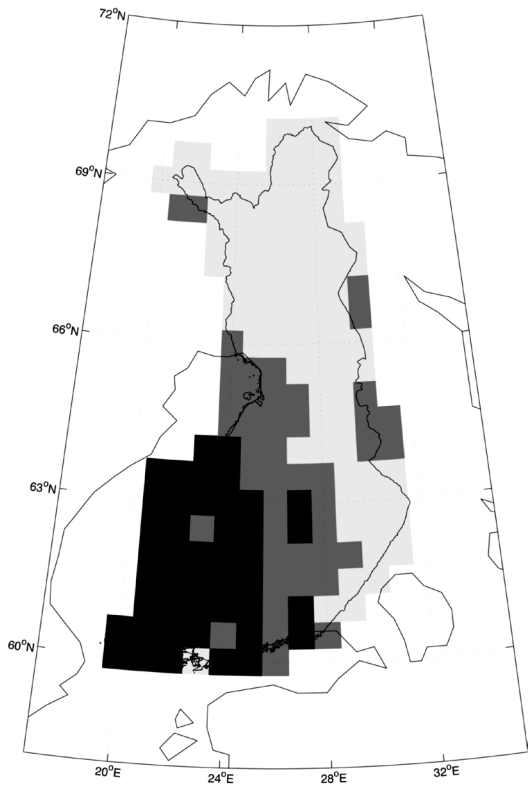

Fig. 6. Regional distribution of the ^{125}Sb deposition (Bq m^{-2}) in Finland decay-corrected to 1 May 1986. The three classes of deposition represent the 0–50, 50–75, and 75–100 percentiles of the interpolated deposition values. The three classes of deposition represent the 0–50 (light grey, < 96 Bq m^{-2}), 50–75 (dark grey, 96–330 Bq m^{-2}) and 75–100 (black, > 330 Bq m^{-2}) percentiles of the interpolated deposition values.

ments. This has been explained by the formation of volatile ruthenium oxides. Ruthenium tetroxide has a melting point of 25.5 °C (Devell *et al*. 1986, Jantunen *et al*. 1991). The high deposition of ^{106}Ru at the Iisalmi–Kuopio region, Kuhmo region and Kotka–Kouvola region (Fig. 5) is similar to the observed ^{103}Ru deposition (Arvela *et al*. 1990). On the other hand, a zone of a relatively low deposition from Uusikaarlepyy to Lappeenranta was also observed for ^{103}Ru.

The deposition pattern of ^{125}Sb resembled those of cesium isotopes (Fig. 6). Again the areas of the highest deposition were south-western Finland, Kotka–Kouvola region and Varkaus–Kuopio region. The results suggest that antimony, having a melting point of 631 °C and boiling point of 1750 °C, behaved like volatile nuclides in the destroyed reactor. For comparison, in Greece the ^{125}Sb deposition varied between 0 and 4.6 kBq m^{-2} (National Technical University of Athens: http://arcas.nuclear.ntua.gr).

An estimation of the total activity of the five studied nuclides deposited in Finland was obtained by multiplying the arithmetic means of the interpolated deposition values (Table 5) with the geographical area of Finland, 338 000 km^2. The results are presented in Table 6, which includes also a comparison to the reactor core inventory and total emissions (International Atomic Energy Agency 1986, USSR State Committee on the Utilization of Atomic Energy 1986). Of the five studied gamma emitters, ^{137}Cs

Table 6. Total activity deposited in Finland (decay-corrected to 1 May 1986) and fractions from core inventory and atmospheric releases (International Atomic Energy Agency 1986, USSR State Committee on the Utilization of Atomic Energy 1986).

Nuclide	Total deposition (Bq) (1 May 1986)	Total deposition (g) (1 May 1986)	Fraction of inventory (%)	Fraction of emissions (%)
Cs-134	2.5×10^{15}	52	1.3	13
Cs-137	4.3×10^{15}	1400	1.5	12
Ru-106	1.4×10^{15}	12	0.071	2.4
Sb-125	1.1×10^{14}	2.8		
Ce-144	5.5×10^{14}	4.6	0.017	0.61

was found to be the most abundant nuclide both by activity and mass in the Chernobyl-derived fallout in Finland. Its total deposition value obtained in this study, 4.3 PBq, is somewhat higher than the value reported by De Cort *et al*. (1998) which was 3.1 PBq. This discrepancy is partly explained by the reason that, contrary to the other nuclides in this study, the ^{137}Cs estimate contains an unknown fraction originating from the atmospheric nuclear tests. According to De Cort *et al*. (1998) Finland received the highest amount of ^{137}Cs from the accident outside the former Soviet Union. Depending on the nuclide, 0.017%–1.5% of the reactor core inventory and 0.6%–13% of the atmospheric emissions were deposited in Finland. The spatial unevenness of the Chernobyl deposition can be observed also from the large differences between the arithmetic and the geometric means of the interpolated deposition values in Table 5.

Conclusions

This study verifies the regional deposition pattern of ^{137}Cs obtained with aircraft and carborne *in-situ* measurements. The results include the first nation-wide deposition estimates of ^{134}Cs, ^{144}Ce, ^{106}Ru and ^{125}Sb. The behaviour of ^{125}Sb was found to be similar to that of cesium during the release and transport of contamination from the destroyed reactor. It was also noted that depending on the nuclide 0.017%–1.5% of the reactor core inventory and 0.6%–13% of the atmospheric emissions were deposited in Finland. These percentages were proportional to the volatility of the nuclides. In the future the data reported here can be used in the verification of long-range atmospheric transport and dispersion models such as SILAM (Sofiev *et al*. 2006).

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